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16. ABSTRACT The paper discusses historic emissions of volatile organic compounds (VOCs) and nitrogen oxides (NOx) in the U.S. from 1900 to 1985. Total VOC emissions have increased over the study period with peaks around 1930 and 1970. In 1900, transportation sources contributed 2% of the total emissions. By 1960, these sources contributed nearly 50% of the total VOC emissions. Also, the percentage contribution of industrial processes increased from about 3% in 1900 to 26% in 1960 and 39% in 1980. External fuel combustion was a major category from 1900 to 1940, but the percentage contribution has decreased since then. Similarly, the contribution by miscellaneous other sources has also decreased with time. The percentage contribution of solid waste disposal appears to have remained nearly constant over time. NOx emissions have increased nearly steadily since 1900, while SO2 emissions have gradually increased from 1900 to 1980 with considerable changes during certain shorter periods of time. VOCs have also increased, but the rate of increase appears to be less than that of NOx and more than SO2, at least on the national level.

17. KEY WORDS AND DOCUMENT ANALYSIS

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Pollution	Pollution Control	13B
Emission	Stationary Sources	14G
Organic Compounds	Volatile Organic Compounds (VOCs)	07C
Volatility		20M
Sulfur Dioxide		07B
Nitrogen Oxides		

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HISTORIC EMISSIONS OF VOC AND NO_x IN THE UNITED STATES FROM 1900 TO 1985

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1. INTRODUCTION

Volatile organic compounds (VOC) and oxides of nitrogen (NO_x) are considered important to the formation of photochemical smog and ozone. While sulfur dioxide (SO_2) and NO_x have long been considered primary precursors of acid precipitation, research is showing that VOC is also an important constituent in many atmospheric chemical reactions. The historic trend of VOC emissions is important to understanding the long term trend in ozone and smog formation.

2. METHOD

Annual estimates of emissions of VOC are presented for each of the contiguous 48 states and the District of Columbia. Emissions were estimated using a detailed methodology for every fifth year from 1900 to 1985. The state level and the 5-year interval were selected to produce estimates comparable to the historic estimates of SO_2 and NO_x provided by Gschwandtner et al.[1] The VOC emissions are consistent in methodology from year to year and state to state. In addition, the total VOC emissions of all states are identical to the national emissions reported by the U.S. EPA for common years in the period from 1940 to 1985.[2] Annual emissions were estimated by apportioning the national estimates for every fifth year according to yearly activity indicators representing the major source categories.[3]

The major source categories are:

1. Transportation sources
2. External fuel combustion sources
3. Industrial processes
4. Solid vaste disposal
5. Miscellaneous other sources

Values were calculated by aggregating the emissions estimated for smaller categories comprising each major category. These subcategories represent sources for which emission factors and activity indicators could be obtained directly from the U.S. EPA national trends estimates.[2] Collectively, these source categories are considered to account for all anthropogenic emissions.

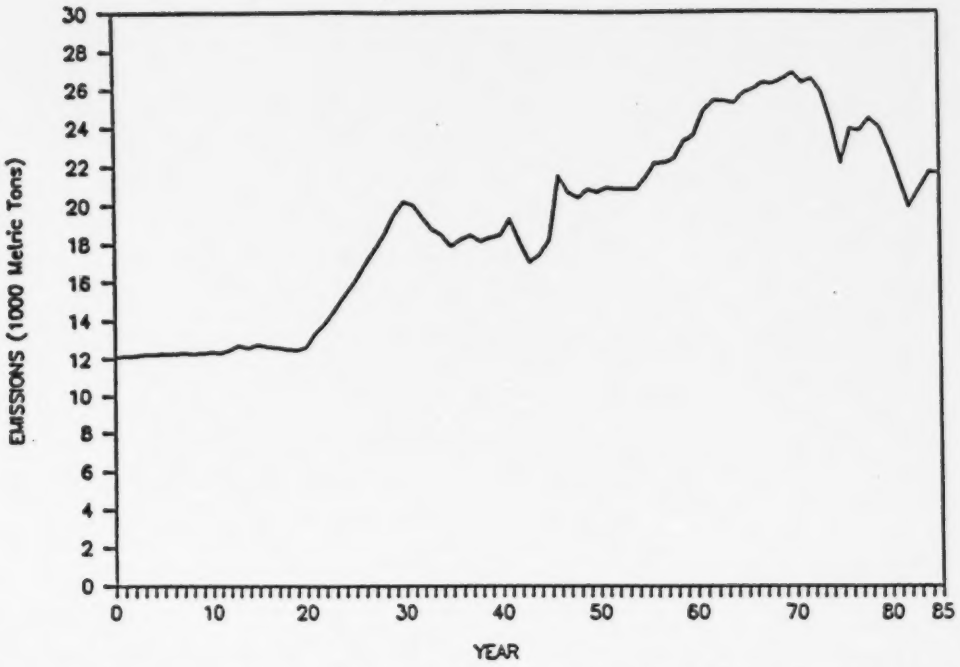

Figure 1. Total VOC emission trend for the United States from 1900 to 1985.

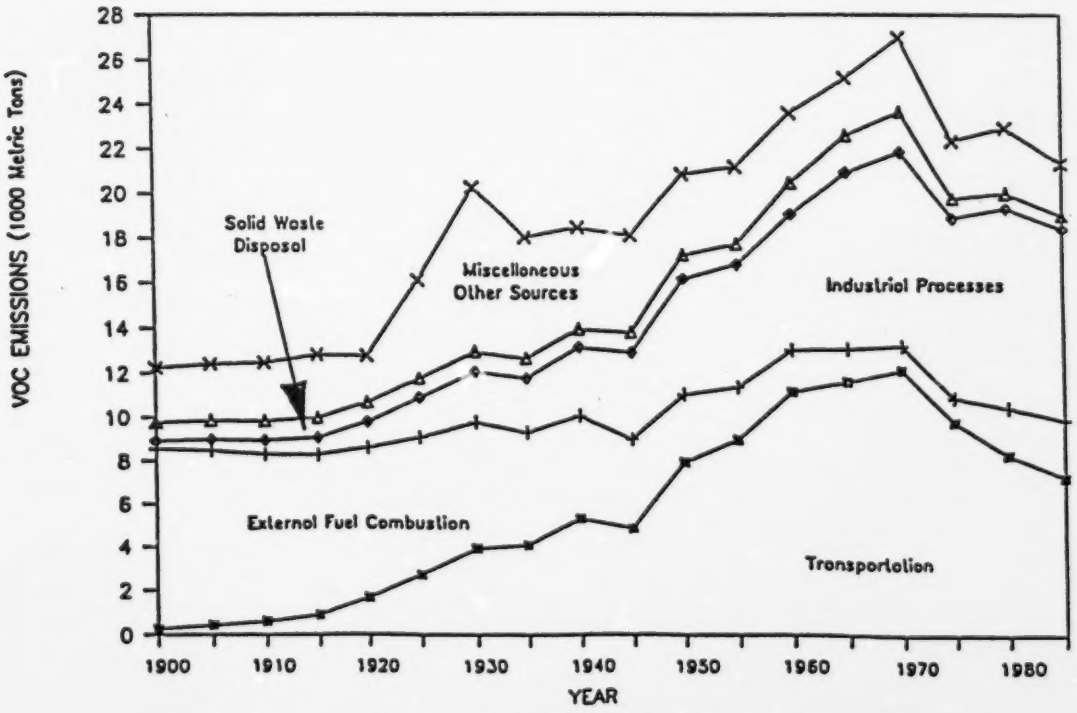

Figure 2. Overall trend in VOC emissions by source category.

3. RESULTS

Figure 1 shows the total VOC emissions for the contiguous United States estimated for each study year and interpolated for the intervening years. The total VOC emissions have increased over the study period with peaks around 1930 and 1970. In 1930, the emissions were higher due mostly to a larger-than-usual number of wildfires, especially in the southeastern and south central states. By 1970, emissions had increased as a result of generally greater activity, especially in the transportation and industrial process categories. After 1970, the total VOC emissions appear to decrease primarily as a result of improved emission control efficiencies on automobiles and many industrial processes, as reflected in the emission factors derived for many of these source categories.

In 1900, transportation sources contributed 2 percent of the total emissions. By 1960, these sources contributed nearly half of the total VOC emissions. Also, the percentage contribution of industrial processes increased from about 3 percent in 1900 to 26 percent in 1960 and 39 percent in 1980. External fuel combustion was a major category from 1900 to 1940, but the percentage contribution has decreased since then. Similarly, the contribution by miscellaneous other sources has also decreased with time, primarily due to the success of fire prevention programs. The percentage contribution of solid waste disposal appears to have remained nearly constant over time.

Figure 2 shows the overall trend in VOC emissions from each major category for the contiguous United States. The figure shows the declining contribution of external fuel combustion sources and the increasing contribution of transportation sources and industrial processes. In the 1920s and 1930s, wildfires contributed significantly to the total VOC emissions which results in the intermediate peak. The contribution of wildfires may also have been significant in earlier years, although exact statistics are not available. From 1970 to 1985, the emissions of many source categories decreased, despite increased activity, as a result of implementing emission controls as reflected in the estimated emission factors.

4. COMPARISON TO SO_2 AND NO_x EMISSIONS

Figure 3 shows the total VOC emissions estimated in this study and the total yearly SO_2 and NO_x emissions estimated by Gschwandtner et al. for 1900 to 1980.[1] The figure shows that the emissions of each of these pollutants have changed over time, but to different degrees. NO_x emissions have increased nearly steadily since 1900, while SO_2 emissions have gradually increased from 1900 to 1980 with considerable changes during certain shorter periods of time. VOC emissions have also increased, but the rate of increase appears to be less than that of NO_x and more than SO_2, at least on the national level.

Since the 1940s, transportation and industrial process combined have contributed most of the VOC emissions. Transportation sources are also major contributors of NO_x. The ratio of VOC to NO_x emissions on the national scale

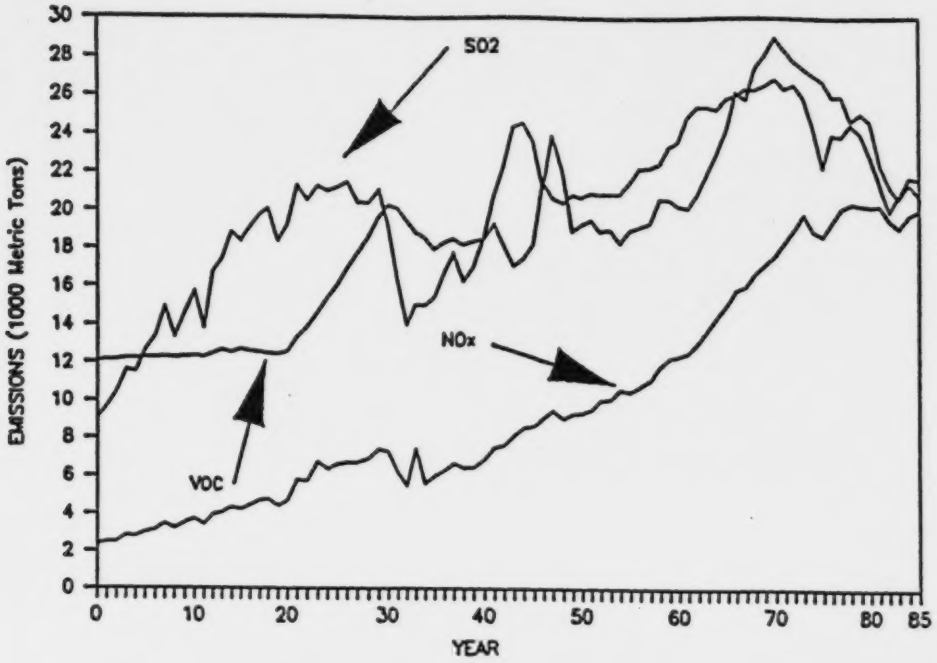

Figure 3. Historic VOC, SO_2, and NO_x emissions for the contiguous United States.

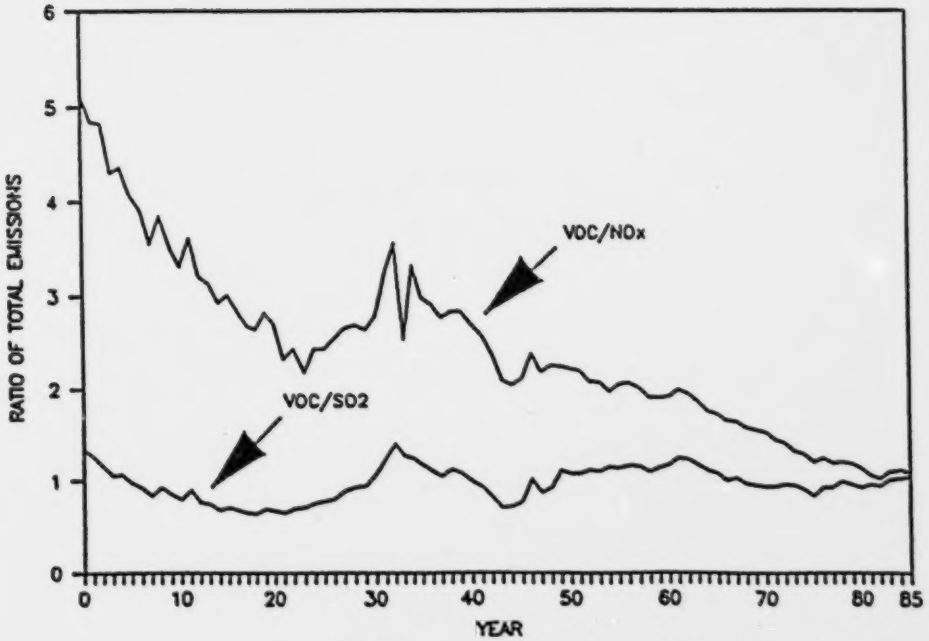

Figure 4. Ratio of VOC emissions to SO_2 and NO_x emissions from 1900 to 1985.

has decreased from 5.1 (based as the median VOC emission estimate in 1900) to 1.1 in 1980, despite an increase in the total emissions of each pollutant. The ratio of VOC to NO_x is an important tool in modeling photochemical pollution such as ozone and smog, and in determining emission reductions necessary to attain national ambient standards for ozone.

For states such as Oregon, Washington, and Idaho where wood burning was the predominant source, the VOC to NO_x ratio has decreased from about 20 to 1. For industrial states such as Nev York, Ohio, and Illinois, the ratio has changed the least due to a mix of predominant sources that collectively emit VOC and NO_x in comparable amounts.

The ratio of VOC to SO_2, however, has remained about equal to unity over the study period, even though these pollutants are emitted by different sources and to different degrees. Figure 4 shovs graphically the historic ratios of VOC emissions to SO_2 and NO_x emissions.

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